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This is the final report on the research performed under AFOSR grant F49620-02-0059. In conjunction with another AFOSR grant (F49620-02-0057), we undertook a comprehensive research program to address the fundamental issues of optical refrigeration in solids important for the development of practical solid-state cryocoolers. We pursued two main avenues of research: First, we explored new rare-earth-doped glasses and crystals that have improved cooling efficiencies and/or lower operating temperatures. This phase also investigated the fundamental physics of optical refrigeration as well as limitations imposed by the structure and composition of the glass for reaching cryogenic temperatures. Second, we studied semiconductor-based optical refrigerators that are much more compact and may achieve appreciably lower operating temperatures than devices based on rare-earth-doped glasses or crystals.

I. Optical Refrigeration in Rare-Earth Doped Materials:

Records have been set in laser cooling of Yb-doped glass. New benchmarks have been established for: i) lowest temperature, ii) temperature differential relative to ambient, and iii) cooling power (i.e. heat lift).

A diode-pumped Yb:YAG disk laser generates 9.6 Watts of power. This laser pumps a specially prepared ZBLAN glass sample doped with 2% Yb in a multi-pass geometry. Care is taken to cut the sample without cracking; all surfaces are then polished to minimize light scattering. The sample is placed in a shell structure that greatly reduces radiative heat loading from the walls of the vacuum chamber. The sample is also isolated conductively from the chamber by mounting it on pedestals fashioned from optical fiber. Three thermocouples monitor temperature on the shell housing, the cold finger to which the shell is attached, and the sample.

Fig. 1 shows results of the record cooling run. The laser is turned on at $t = 0$; steady state cooling is reached in approximately 2 hours. Note the upward spike of housing temperature (blue and green curves) that occurs when it is exposed to scattered laser light. The sample thermocouple reading (red solid curve) is also affected by fluorescence. This is clearly evident when the pump laser is turned off at $t = 3$ hours and the measured temperature immediately drops. A corrected sample temperature (red dotted curve) corrects for this thermocouple error.

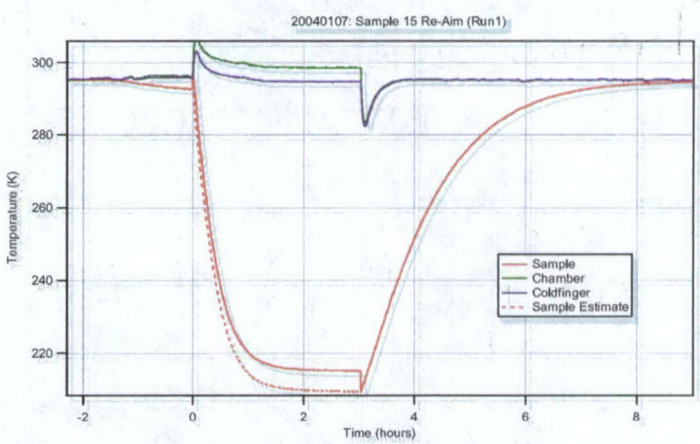

Fig.1 Laser cooling of Yb-doped ZBLAN glass by an amount 88 K. Illumination commences at $t = 0$ and steady-state is reached after about 3 hours. The upper curves depict thermocouple monitors of the chamber temperature, which is slightly heated by the escaping luminescence. The dotted curve is the deduced sample temperature when corrected for fluorescence heating of the thermocouples.

The lowest temperature observed is 208 K and the maximum differential between the housing and sample is

88 K. This minimum represents a 30 degree improvement over the previous best result. It also demonstrates that laser cooling of solids can provide temperature drops that are comparable with current thermoelectric coolers. The estimated cooling power in this experiment is 45 mW, which is approximately two times better than the previous record.

We made the first demonstration of laser cooling in a new host: BaY_2F_8 (see Fig. 2). This material is interesting for several reasons including its potential for higher efficiency compared to ZBLAN due to lower phonon energy, its infrared transparency making it less susceptible to radiative heat loading, and higher relative hardness leading to better polished surfaces. The sample is doped with 2% Tm and pumped in the spectral region around 1.9 um in a single pass 'proof-of-principle' arrangement. Cooling by an amount 1.5 degrees below ambient is obtained for 6 Watts of pump power. Preliminary studies also indicate that the background absorption in this material is much lower than Tm:ZBLAN, making it quite attractive for achieving cryogenic temperatures. Our collaborators at INFN in Pisa, Italy have grown Yb-doped BaY_2F_8. We obtained laser cooling in this crystal for the first time in experiments with YAG laser pumping.

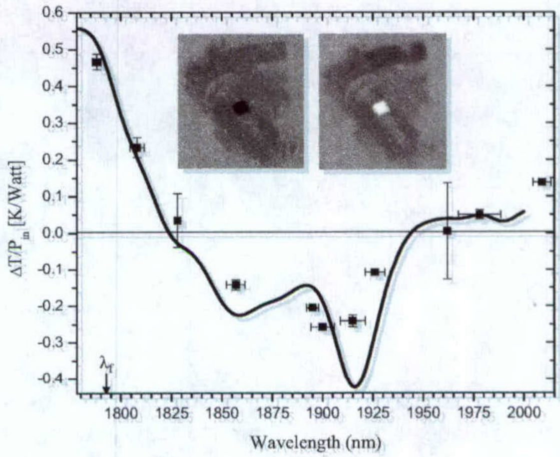

Fig. 2 Temperature change normalized to incident power as a function of pump wavelength for a 2 wt.% $Tm^{3+}:BaY_2F_8$ sample. The solid line is a theoretical fit. The inset shows false color thermal images corresponding to heating at 1788 nm (left photo) and cooling at 1902 nm (right photo).

We explored the concept of resonant cavity pumping (see Fig. 3). The essential idea is to place the cooling element inside a Fabry-Perot resonator; the resonator length is tuned automatically by the servo loop to maximize the absorbed laser power. An ideal system can attain 100% absorption of pump light in theory. There are several keys to this concept: 1) attaining the highest possible reflectivity with the super mirror, 2) designing the proper front mirror reflectivity, and 3) matching the Fabry-Perot resonator phase front curvature to the propagating laser mode. Our most recent proof-of-principle measurements indicate the trapped light is making ~ 15 round trips. Only small improvements in alignment can lead to tenfold or more increase in the number of round trips.

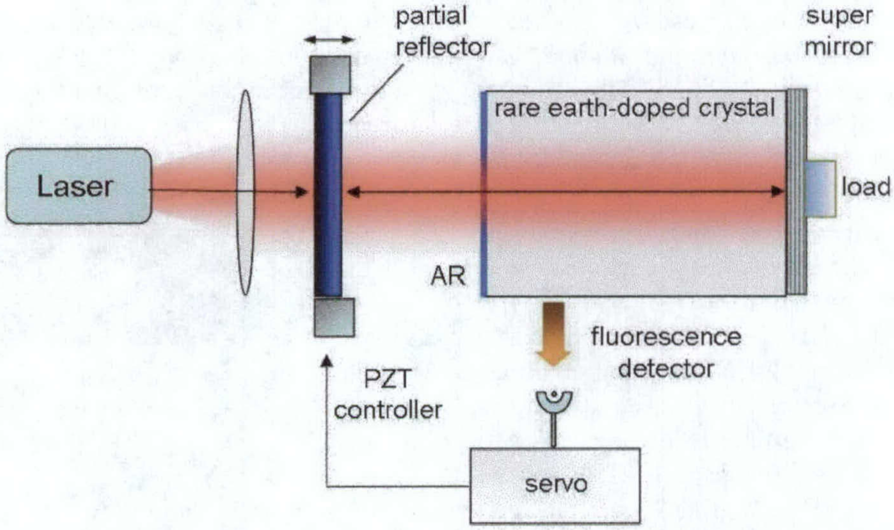

Fig. 3. A laser cooling crystal is placed inside a Fabry-Perot resonator to enhance pump light absorption. We have made preliminary characterization tests of this scheme.

II. Progress on Optical Refrigeration in Semiconductors:

1) *Theory*. We conducted both theoretical and experimental investigations of laser cooling in semiconductors. Our theoretical work brought together the critical issues of recombination lifetimes and the effects of many-body (Coulomb) interactions at low temperatures. This breakthrough research was published in *Physical Review Letters*; this paper discusses the essential issues of laser cooling in semiconductor materials. The key criterion for achieving net cooling in semiconductors defines a lower limit for the non-radiative lifetime τ_{nr}. We require a non-radiative lifetime given by the following inequality:

$$\tau_{nr} > \left(\frac{h\nu}{kT}\right)^2 \frac{4C}{(\eta_e B)^2}$$

where hν is the photon energy, kT the thermal energy, C is the Auger coefficient, B is the radiative recombination coefficient, and η_e the luminescence extraction efficiency. This criterion is plotted in Fig. 4 for GaAs at room temperature. It indicates that for $\eta_e \approx 0.2$ (achievable using a ZnSe dome), a nonradiative lifetime longer than 30 μs is required. We therefore investigated nonradiative lifetimes on various GaAs heterostructures as a function of temperature and thickness.

2) *Time resolved spectroscopy to determine the nonradiative lifetimes*

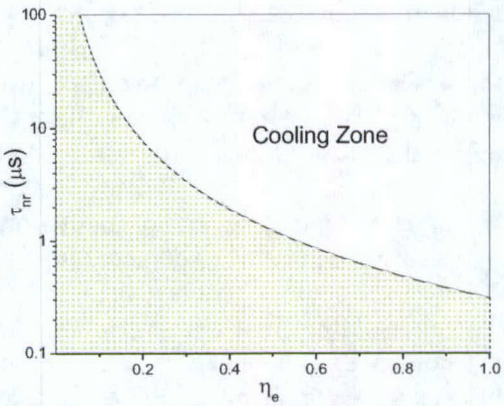

Fig.4 *The break-even nonradiative lifetime plotted versus the luminescence escape efficiency.*

Nonradiative decay in a semiconductor structure is an undesirable process that produces heat. Its origin is primarily surface states and to a lesser extent from impurities. This quantity varies greatly from sample to sample (in our case GaAs) depending on the growth technique and environment as well as heterostructure composition. Our theory indicates that a lifetime of 10 μs or longer is needed for achieving net cooling. We set up a time-resolved photoluminescence (TRPL) experiment as shown in Fig.5. Various GaAs heterostructures purchased from commercial vendors or obtained from our collaborators at national labs were evaluated.

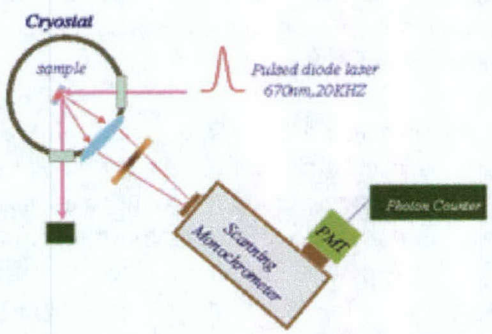

Fig. 5*. Experimental set up for time-resolved photoluminescence (TRPL) studies.*

We also investigated the dependence of the lifetime on epitaxial lift-off. To produce a cooling device, the active heterostructure must be removed from its substrate and then bond it to the ZnS or ZnSe dome using van der Waals mechanism. The result is summarized in Fig.6 and indicates that lift-off and bonding shortens the lifetime by a nearly a factor of two. These results were published in *Applied Physics Letters*. We are currently developing new methods of lift-off and bonding to eliminate such interface degradation.

3) *Differential Luminescence Thermometry*

One of the most challenging problems in laser cooling is obtaining accurate temperature without a deleterious perturbation to the optical cooling element. Thermocouples, for example, may provide a heat conduction path to the ambient. The heat conduction problem can usually be overcome with proper wire routing. More important is absorption of luminescence that accompanies laser cooling. This so-called 'photon waste' can be absorbed by the thermocouple, which then heats up. The thermocouple is in direct contact with the sample and heats it, thereby reducing or eliminating net cooling. Clearly, a non-contact temperature measurement scheme is highly desirable.

We developed a highly sensitive, non-invasive spectroscopic technique for measuring temperature in a semiconductor cooling experiment. It exploits changes that occur in the direct bandgap recombination of GaAs as a function of temperature. Differential luminescence in the vicinity of the GaAs bandgap wavelength provides us with temperature resolution of < 0.05 K.

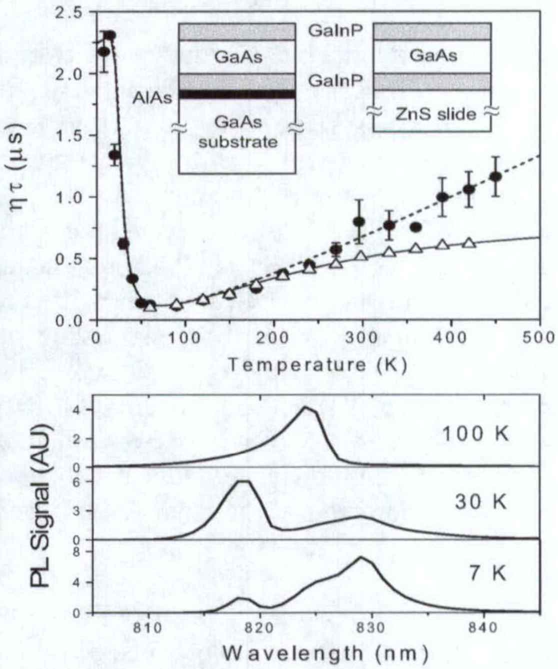

Fig. 6 Photoluminescence lifetime of GaInP/GaAs heterostructures before liftoff (filled circles) and after lift-off and bonding to a ZnS slide (triangles). The dotted curve is from a model that ignores interface recombination; solid curve includes interface recombination. Bottom: Time-integrated photoluminescence of the unprocessed sample revealing acceptor freeze-out at T ~ 30 K.

Implementation of differential luminescence to measure temperature is depicted in Fig. 7. When the sample cools, the bandgap expands resulting in a blueshift of the peak luminescence. Conversely, heating causes a luminescence redshift. Two spectra obtained at slightly different temperatures are superimposed in Fig. 7a and appear identical. After normalization and subtraction, however, a peak and valley are evident as shown in Fig. 7b. The height of the differential peak indicates a temperature change of $\Delta T \approx 0.25$ K.

4) *Time-resolved Luminescence.* In our spectroscopic study of GaAs published in *Applied Physics Letters*, we used a slow-scan grating monochrometer to obtain time-integrated spectra. We have since modified the experiment to obtain full spectra with millisecond temporal resolution. We can now monitor the dynamics of laser cooling in real-time.

The heart of the new system is a time-gated CCD detector. The CCD captures the entire luminescence spectrum in a single shot. We obtain temporal gating as short as 10 ms using a mechanical shutter. Temporal gating three orders of magnitude faster is possible with a high speed intensifier, although higher speed comes at the expense of sensitivity. The CCD detector produced the data shown in Fig. 7. It is important to note that bandgap luminescence data cannot be acquired when high power pump light is present. We disable the pump beam during the luminescence acquisition period with a chopper wheel inside the laser resonator. The pump laser duty cycle is set very low (pump is on ~ 90% of the time). In this way, we can monitor the dynamics of a laser cooling experiment in the presence of a quasi-cw pump beam.

We have used the time-resolved luminescence system to study local cooling of GaAs heterostructures bonded to ZnS domes. No net or local cooling has been detected yet, but we have discovered the crucial importance of photon waste management in the design of our semiconductor cooling experiments. We observe reduced heating when extreme care is used to remove cooling luminescence from the vacuum cryostat housing the sample.

Wavelength dependent heating of our samples allows us to deduce the external quantum efficiency. We are interested in both internal and external quantum efficiencies associated with our devices. Internal quantum efficiency describes how effectively pump laser light gets converted to luminescence. External quantum efficiency is a measure of how well this luminescence escapes the device. External quantum efficiency must always be less than internal. This subject has been recently quantified as we describe below.

Fig. 8 shows the experimental external quantum efficiency for various thickness GaAs structures bonded to a ZnS dome. A clear peak efficiency of 96% occurs at thickness near 0.75 µm. An optimal thickness exists because of the following tradeoff: thin layers increase the probability a photon will escape before being absorbed, but decrease the non-radiative lifetime due to surface recombination. Non-radiative recombination is highly undesirable because it converts optical excitation directly into heat. Our data confirms previous measurements of external quantum efficiency for similar heterostructures.

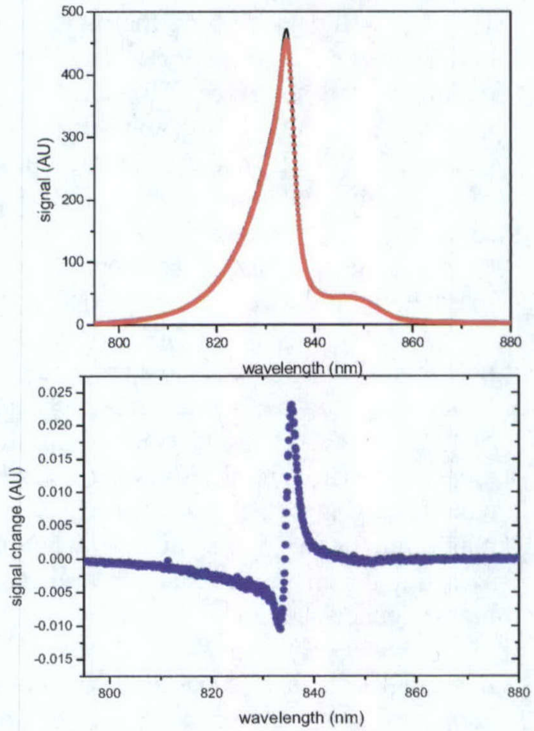

Fig 7. Bandgap luminescence of GaAs at two slightly different temperatures near 150 K. The overlapped spectra (top) appear identical except at the peak. Subtracted spectra (bottom) clearly reveal a difference signal corresponding to a temperature change of 0.25 K.

5) *Analysis of laser cooling in quantum wells.* Our current experimental work has made us keenly aware of the crucial importance of luminescence removal in a semiconductor cooling experiment. The problem with semiconductors when compared to glasses and transparent crystals is the presence of a significant refractive index mismatch at the semiconductor-vacuum interface. This leads to the condition of total internal reflection for the vast majority of cooling luminescence. Trapped luminescence is absorbed and converted to heat, which will likely prevent the realization of net cooling.

With this in mind, we have critically re-examined the only published claim of local laser cooling in a semiconductor quantum well (Finkeissen et al, *Applied Physics Letters*, 1999). In particular, this experiment did not address the total internal reflection problem and luminescence management in general. This deficiency could only result in undesired optical absorption and certain sample heating.

The experimental arrangement used by Finkeissen et al consists of three 9-nm wide quantum wells are situated on a GaAs substrate held at 45 K in a cryostat. The quantum wells are pumped by a cw Ti:sapphire laser at an irradiance of 4 W/cm^2. Because the wells are so narrow, their absorption is very small – essentially all the pump light is absorbed in the GaAs substrate immediately adjacent to the wells. A typical substrate height (> 100 μm) will provide for a thermal gradient between the coldfinger and substrate-quantum well interface. In addition, the majority of luminescence coming from the wells will end up in the substrate because of total internal reflection. The GaAs substrate will heat and transfer thermal energy to the heterostructure convectively, overwhelming local cooling.

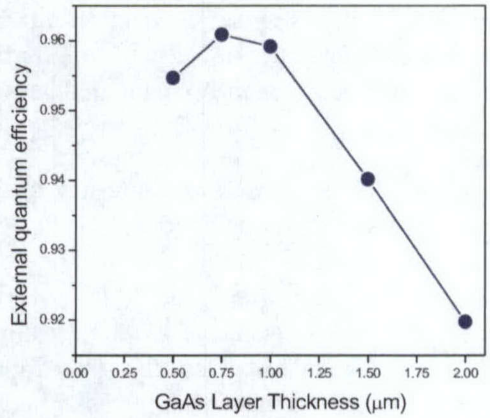

Fig. 8. Measured external quantum efficiency for five different GaAs structures bonded to a ZnS dome at 200 K.

In this experiment, temperature was deduced by monitoring the relative change of luminescence coming from excitonic emission lines in the quantum wells. The calibration measurements were all done at low laser irradiance (5 mW/cm^2) where the excited electron-hole pair density is small. The cooling experiments were performed with an irradiance 3 orders of magnitude higher, i.e. a situation where density effects are no longer negligible. We believe that a carrier density phenomenon – not laser cooling – can explain the observed behavior in the luminescence spectra.

It is well known that the exciton potential is reduced or screened when free carriers have an increasing statistical probability of existing between the bound electron-hole. This could explain the reduced luminescence seen from the exciton line associated with the larger radius orbit. We have modeled 3-D exciton luminescence using the established Banyai-Koch theory and observed changes to the luminescence that are qualitatively consistent with Finkeissen et al.

The rate of bimolecular, radiative recombination increases with increasing excitation. In steady state, radiative recombination scales as the square root of the pump irradiance ($\sqrt{I}$). If the phonon absorption rate is comparable to or slower than radiative recombination, the differential exciton luminescence signal (used to monitor temperature by Finkeissen et al) would be altered. This 'phonon absorption bottleneck' could lead one to incorrectly conclude that cooling exists. We have estimated this rate for the quantum well system and indeed find scatter rates that are plausible for energy levels separated by less than an optical phonon energy.

In conclusion, we report exciting new developments in laser cooling of both glasses and semiconductors. A record amount of temperature decrease was obtained with Yb:ZBLAN. The temperatures we achieved prove that this technology can compete with established thermoelectric devices. A new crystalline host (BaY_2F_8) was cooled for the first time; this material exhibits excellent infrared transparency compared to ZBLAN that could lead to a more efficient and fieldable laser cooler. A comprehensive study of laser cooling in semiconductors uncovered the critical design issues needed to build a working device. We have significantly improved our spectroscopic capabilities for characterizing cooling in semiconductors and demonstrated a powerful, non-contact technique for measuring temperature with high accuracy in real-time. In addition, we have critically studied the only report of laser cooling in a semiconductor and found this claim to be implausible. We understand the physics and engineering problems in semiconductor cooling and are vigorously pursuing an intensive research program to make the first demonstration of net laser cooling in semiconductors.

III. Publications Resulting From This Award:

Journal Publications:

1. B. Imangholi, M. P. Hasselbeck, and M. Sheik-Bahae, "Absorption Spectra of Wide Gap Semiconductors in the Transparency Region" Opt. Comm., **227**, 337 (2003).

2. M. Sheik-Bahae and R. I. Epstein, "Can Laser Light Cool Semiconductors", Phys. Rev. Lett., **92**, 247403 (2004).

3. B. Imangholi, M.P. Hasselbeck, M. Sheik-Bahae,R. I. Epstein, S. Kurtz, "Effects of epitaxial lift-off on interface recombination and laser cooling in GaInP/GaAs heterostructures" Appl. Phys. Lett. **86**, 81104 (2005)

4. J. Thiede, J. Distel, S. Greenfield, R. Epstein, "Cooling to 208 K by optical refrigeration", Appl. Phys. Lett., **86**, 154107 (2005).

Refereed Conference Presentations

1. **(INVITED)** M. P. Hasselbeck, M. Sheik-Bahae, J. Thiede, J. Distel, S. Greenfield, W. Patterson, S. Bigotta, B. Imangholi, D. Seletskiy, D. Bender, V. Vankipuram, N. Vadiee, and R. I. Epstein, *"Laser Cooling of Infrared Sensors*" SPIE 49th Annual Meeting, Denver, Colorado, Aug. 2004.

2. B. Imangholi, M. P. Hasselbeck, and M. Sheik-Bahae. R. I Epstein, S. Kurtz, "Laser Cooling in Semiconductors: Is it possible?" *International Quantum Electronics Conference (IQEC),* paper IMO-6, San Francisco, CA, May 2004, OSA

3. W. Patterson, M. P. Hasselbeck, M. Sheik-Bahae, S. Bigotta, M. Tonelli, R. I. Epstein, J. Thiede, "Observation of optical refrigeration in Tm^{+3}:BaY_2F_8," *Conference on Lasers and Electro Optics (CLEO),* paper CThE-5, San Francisco, CA, May 2004, OSA.

4. M. Sheik-Bahae, B. Imangholi, M. P. Hasselbeck, R. I Epstein, S. Kurtz, "Nonlinear Optical Processes in Laser Cooling of Semiconductors," NLO conference, Hawaii, Aug. 2004.

5. W. M. Patterson, A. Mocofanescu, M. Sheik-Bahae, J. Thiede, R. Epstein, S. Bigotta, D. Parisi, A. Toncelli, M. Tonelli, *Laser cooling in rare earth doped* BaY_2F_8 *crystals*, submitted to the 2006 OSA Annual Conference.